

Article

Pyrometallurgical Recovery of Neodymium from Nd–Fe–B Magnets in End-of-Life Electric Vehicle Motors Using Non-Ferrous Smelting Slag Flux

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Abstract

The increasing use of electric vehicles has led to a growing demand for rare-earth elements, particularly neodymium (Nd), which is a critical component of Nd–Fe–B permanent magnets used in traction motors. End-of-life electric vehicle motors are therefore considered promising secondary resources for Nd recovery. In this study, a pyrometallurgical process using non-ferrous smelting slag as a flux was proposed for recovering Nd from Nd–Fe–B magnets contained in waste electric vehicle motors. Steel and magnet fractions obtained from a dismantled motor were melted at approximately 1600 °C under an air atmosphere, and Fe₂O₃ was added as an oxidizing agent to promote the selective oxidation of Nd. The oxidized Nd was subsequently partitioned into the slag phase as Nd₂O₃ through metal–slag separation. The effects of the slag flux addition ratio, Fe₂O₃ content, slag flux type, and crucible material on Nd recovery behavior were systematically investigated. Increasing the fayalite-based slag flux addition enhanced Nd transfer into the slag phase, and the highest Nd recovery of approximately 80% was obtained at a slag flux addition ratio of 30 wt%. The addition of 2 wt% Fe₂O₃ was found to be suitable for promoting stable Nd oxidation and efficient slag–metal separation. The non-ferrous smelting slag exhibited chemical and phase characteristics comparable to those of synthetic fayalite slag flux, resulting in similar Nd recovery performance. In addition, the crucible material significantly affected the process stability and Nd recovery behavior. Although a carbon crucible showed relatively high Nd recovery, severe slag foaming and crucible erosion occurred during melting. In contrast, alumina and MgO crucibles provided stable process conditions with comparable Nd recovery behavior, whereas zirconia crucibles caused relatively higher Nd loss. These results demonstrate that non-ferrous smelting slag can be effectively used as a flux for the pyrometallurgical recovery of Nd from end-of-life electric vehicle motors, offering a potential route for sustainable rare-earth recycling.

Keywords: neodymium recovery; Nd–Fe–B magnets; end-of-life electric vehicle motors; non-ferrous smelting slag; pyrometallurgical recycling

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1. Introduction

The global transition toward carbon neutrality has accelerated the shift from fossil-fuel-based transportation systems to electrified mobility. Electric vehicles (EVs) have received considerable attention as a key technology for reducing greenhouse gas emissions

and improving energy efficiency in the transport sector. Supported by government policies and industrial investment, the global EV market has expanded rapidly and is expected to continue growing over the coming decades [1]. This growth has also increased the demand for critical raw materials required for EV powertrains, batteries, and related components [2].

Among the key components of EV traction systems, permanent-magnet motors are widely used because of their high power density, compact design, and high energy-conversion efficiency [3]. In particular, neodymium–iron–boron (Nd–Fe–B) permanent magnets are essential for high-performance EV motors, wind turbines, and various advanced electrical devices. Neodymium (Nd) plays a central role in providing high magnetization and coercivity, enabling miniaturization and efficiency improvements in motor systems [4]. However, the supply of rare-earth elements, including Nd, is highly concentrated in a limited number of countries, making the global supply chain vulnerable to geopolitical risks, export restrictions, and price fluctuations [5].

Because many countries, including Korea, rely heavily on imports of rare-earth resources, securing secondary resources has become increasingly important. Urban mining, which involves the recovery of valuable metals from end-of-life products and industrial waste streams, has emerged as an important strategy for reducing dependence on primary resources and improving resource circularity [6]. With the rapid increase in EV deployment, the amount of end-of-life EV motors is expected to increase substantially, and these motors can serve as promising secondary resources for Nd recovery [7]. EV motors consist of complex assemblies containing Fe, Cu, Al, and other structural metals, while Nd–Fe–B magnets are embedded in the rotor [8]. However, because of the complex structure and strong integration of metallic components and magnets, simple physical separation is often insufficient for efficient Nd recovery.

Hydrometallurgical processes based on acid leaching have been widely investigated for the recovery of rare-earth elements from spent Nd–Fe–B magnets and related wastes [9]. These processes can achieve high extraction efficiency but generally require large amounts of chemical reagents, multi-step separation, and wastewater treatment, resulting in environmental and economic burdens [10]. In the case of large and complex waste streams such as end-of-life EV motors, additional pretreatment steps, including dismantling, crushing, and selective separation, are required before leaching. These steps increase energy consumption and process complexity, limiting the applicability of purely hydrometallurgical routes for large-scale motor recycling [11].

In contrast, pyrometallurgical processing offers several advantages for treating complex and bulky secondary resources. High-temperature smelting can directly process mixed metallic components and promote phase separation based on differences in chemical affinity, density, and oxidation behavior [12]. In metal–slag systems, selective oxidation can be used to transfer target elements from the metal phase to the slag phase. Nd has a much higher affinity for oxygen than Fe and readily forms stable Nd_2O_3 under oxidizing conditions at high temperatures [13]. Therefore, when an appropriate oxidizing agent and slag system are used, Nd can be selectively oxidized and concentrated in the slag phase, while Fe remains mainly in the metallic phase [14].

Slag design is a critical factor in such pyrometallurgical recovery processes. FeO – SiO_2 -based fayalite slag flux has a relatively low melting temperature and suitable fluidity, making it favorable for metal–slag separation and oxide dissolution [15]. The addition of Fe_2O_3 can increase the oxygen potential of the slag–metal system and promote the selective oxidation of Nd into Nd_2O_3 [16]. Recent studies have also emphasized that the development of sustainable rare-earth recycling processes requires not only high recovery efficiency but also the use of practical flux materials, reduced chemical consumption, and improved process scalability [17,18]. In this context, non-ferrous smelting slag is attractive

as an alternative flux because it already contains FeO–SiO₂-based components similar to fayalite slag and can potentially be reused as a functional slag-forming material.

Several studies have reported pyrometallurgical strategies for recovering rare-earth elements from spent Nd–Fe–B magnets, including oxidation, smelting, and slag-based separation approaches [16,19,20]. However, most studies have focused on magnet scrap or pretreated magnet powders, whereas direct treatment of end-of-life motor-derived materials remains less explored. In addition, the use of real non-ferrous smelting slag as a flux for Nd recovery from waste EV motor components requires further investigation, particularly with respect to slag composition, Fe₂O₃ addition, metal–slag separation, and crucible-related reactions.

Therefore, this study proposes a non-ferrous smelting slag flux-based pyrometallurgical process for recovering Nd from Nd–Fe–B magnets contained in end-of-life EV motors. Steel and magnet fractions derived from a waste motor were mixed with FeO–SiO₂-based slag flux and Fe₂O₃ and then melted at approximately 1600 °C under an air atmosphere. The process was designed to induce the selective oxidation of Nd and its partitioning into the slag phase as Nd₂O₃. The effects of slag flux addition ratio, Fe₂O₃ content, slag flux type, and crucible material on Nd distribution and recovery behavior were systematically investigated. Through this approach, the feasibility of using non-ferrous smelting slag as a functional flux for sustainable Nd recovery from end-of-life EV motors was evaluated.

2. Materials and Methods

2.1. Experimental Overview

In this study, a pyrometallurgical process based on metal–slag separation was applied to recover neodymium (Nd) from Nd–Fe–B magnets contained in end-of-life electric vehicle (EV) motors. The main recovery mechanism was designed to selectively oxidize Nd in the molten metal phase and subsequently partition the oxidized Nd into the slag phase. Fe₂O₃ was used as an oxidizing agent to promote the formation of Nd₂O₃, while FeO–SiO₂-based fayalite slag flux was employed as the reference slag system to evaluate the distribution behavior of Nd between the metal and slag phases. The effects of slag flux addition ratio, Fe₂O₃ content, reaction temperature, and slag flux type on Nd recovery behavior were investigated. After determining the optimum conditions using synthetic fayalite slag flux, non-ferrous smelting slag was applied under the same conditions to evaluate its feasibility as an alternative industrial slag flux. In addition, the influence of crucible material on Nd recovery and process stability was examined as a secondary process variable. Through these experiments, the optimum conditions for Nd recovery and the applicability of non-ferrous smelting slag as a functional flux for rare-earth recycling were evaluated.

2.2. Experimental Materials

The end-of-life EV motor used in this study was provided by Hyundai Motor Co. The motor consisted of several components, including the housing, stator, rotor, and Nd–Fe–B magnets. Because the chemical composition and material ratio of EV motors vary depending on the manufacturer and model, the entire motor was not directly used in the smelting experiments. Instead, the rotor part, where the Nd–Fe–B magnets are embedded, was dismantled and used as the main target material for Nd recovery.

The rotor was mainly composed of steel and Nd–Fe–B permanent magnets. Since Nd is concentrated in the magnet fraction, the steel and magnet components were separated from the rotor and weighed to determine the experimental charge composition. The rotor-derived charge consisted of 94.32 wt% steel and 5.68 wt% Nd–Fe–B magnet. The Nd–Fe–

B magnet was assumed to contain 27–32 wt% Nd, 67–73 wt% Fe, and approximately 1 wt% B, based on the typical composition of commercial Nd–Fe–B magnets. Because the experiments were conducted at the laboratory scale, direct melting of the entire rotor assembly was not practical. Therefore, the rotor components were cut, separated, and recombined according to the measured mass ratio of steel and magnet. This approach allowed the actual rotor composition to be simulated while maintaining reproducible laboratory-scale experimental conditions. Based on this composition, steel, Nd–Fe–B magnet, slag flux, and Fe_2O_3 were mixed in predetermined ratios and used as starting materials for the pyrometallurgical Nd recovery experiments.

2.3. Slag Flux and Oxidizing Agent

In this study, FeO–SiO₂-based fayalite slag flux was used as the reference slag system to evaluate the selective oxidation and slag-phase partitioning behavior of Nd. In addition, non-ferrous smelting slag flux generated from an industrial non-ferrous smelting process was applied to examine the practical applicability of the proposed process. Fayalite slag was selected because the FeO–SiO₂ system has a relatively low melting temperature and favorable fluidity at high temperatures. Compared with SiO₂-rich slag, FeO acts as a network modifier and reduces the polymerized silicate structure, thereby improving slag fluidity. These characteristics are advantageous for promoting metal–slag separation and enhancing the dissolution and capture of metal oxides such as Nd₂O₃ in the slag phase. In addition, because fayalite slag contains FeO, it can provide a suitable reaction environment for Fe-based metal–slag systems and selective oxidation reactions. The fayalite slag flux used in this study was synthesized by mixing FeO and SiO₂, followed by a high-temperature reaction. After synthesis, the fayalite slag was crushed and homogenized to improve its reactivity and ensure uniform mixing with the experimental charge. The synthesized fayalite slag flux was used as the reference slag system for evaluating the effects of slag flux addition and Fe_2O_3 addition on Nd recovery behavior.

Non-ferrous smelting slag flux was also used as an alternative slag-forming material. This slag was generated from an actual industrial non-ferrous smelting process and had a fayalite-based composition containing additional oxides such as Al₂O₃, CaO, and MgO. These minor components can affect the viscosity, oxygen potential, oxide solubility, and distribution behavior of Nd between the metal and slag phases. Therefore, after the optimum conditions were determined using the synthesized fayalite slag flux, the non-ferrous smelting slag flux was applied under the same conditions to compare its Nd recovery behavior and evaluate its feasibility as an industrial slag flux. Fe_2O_3 was used as the oxidizing agent. Nd has a high affinity for oxygen and readily forms stable Nd₂O₃ under high-temperature oxidizing conditions. Fe_2O_3 can supply oxygen to promote the selective oxidation of Nd, while the reduced Fe can be incorporated into the metallic phase. Therefore, Fe_2O_3 was considered suitable for this Fe-based smelting system because it can induce Nd oxidation without introducing foreign metallic impurities into the process.

The Fe_2O_3 addition amount was varied from 0 to 4 wt% based on the total mass of the experimental charge to evaluate its effect on Nd oxidation and slag-phase partitioning. The range was selected to include insufficient, optimum, and excess oxidizing-agent conditions. Insufficient Fe_2O_3 addition may limit the oxidation of Nd and reduce its transfer into the slag phase, whereas excessive Fe_2O_3 addition may change the slag composition and oxygen potential, potentially affecting Nd distribution behavior. Therefore, the effect of Fe_2O_3 addition on Nd recovery was systematically investigated within this range. The slag flux addition ratio was set to 10, 20, and 30 wt% based on the total mass of the experimental charge. If the slag flux amount is insufficient, the generated Nd oxide may not be fully dissolved or captured in the slag phase and may remain in the metal phase or at the metal–slag interface. Conversely, excessive slag addition can decrease process efficiency

because of dilution effects and increased slag volume. Therefore, the slag flux addition ratio was varied to evaluate Nd partitioning behavior and determine the optimum condition for Nd recovery.

2.4. Experimental Apparatus

A high-frequency induction furnace (Shinsung Co., Ltd., Gimpo, Republic of Korea) was used for melting the experimental charge and promoting metal–slag separation. The induction furnace system used in this study consisted of a melting chamber, geared motor, power control unit, and chiller unit. The system was equipped with an insulated-gate bipolar transistor (IGBT)-based TR inverter with a maximum power of 50 kW and an operating frequency of 15 kHz. The furnace had a melting capacity of approximately 25–30 kg based on steel and was equipped with both automatic and manual operation modes through a silicon-controlled rectifier (SCR)-based DC control system. A 10 HP chiller system was used to circulate cooling water and prevent overheating of the induction coil and power supply components. The melting chamber was used for charge melting, slag formation, and metal–slag separation. The geared motor controlled the tilting motion of the furnace, enabling stable discharge and recovery of the molten products. The power control unit regulated the operating parameters, including voltage, current, and frequency, while the chiller unit maintained stable cooling of the furnace system during operation. During induction heating, an induced current is generated within the metallic charge by electromagnetic induction. The resulting electromagnetic force induces stirring in the molten metal, which can promote heat transfer, homogenization of the melt, slag formation, and interaction between the metal and slag phases. In addition, thermal convection generated by temperature gradients in the molten bath can further enhance mass transfer and compositional homogenization. These effects are advantageous for inducing efficient oxidation, slag formation, and phase separation during the pyrometallurgical recovery of Nd. In this study, the electromagnetic stirring and thermal convection characteristics of the induction furnace were utilized to promote metal–slag reactions and evaluate the distribution behavior of Nd between the metal and slag phases.

2.5. Experimental Design

Figure 1 shows the overall process flow for Nd recovery from the rotor-derived material. Because the general process concept has already been described in Section 2.1, this section focuses on the experimental variables, thermodynamic calculation conditions, and uncertainty estimation methods used to evaluate Nd distribution and recovery. Briefly, the process consisted of melting the steel and Nd–Fe–B magnet-containing charge at approximately 1600 °C, selective oxidation of Nd using Fe_2O_3 , dissolution of the generated Nd oxide into the slag phase, and final separation into an Fe-rich metallic phase and a rare-earth-element-bearing slag phase.

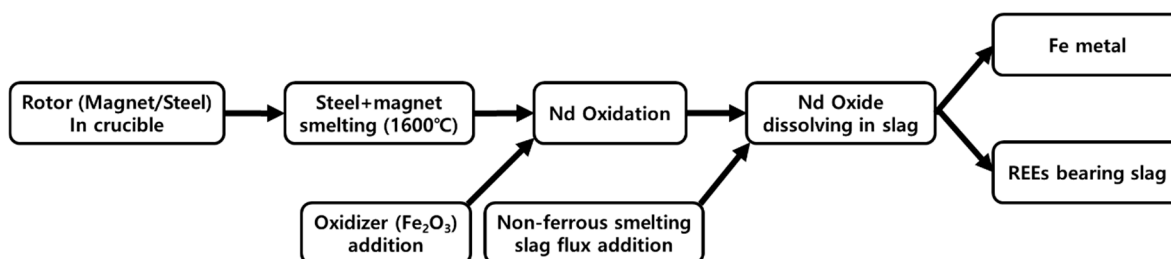


Figure 1. Overall experimental process flow for Nd recovery from waste electric motor.

Thermodynamic calculations were performed using FactSage 8.2 to provide qualitative guidance for selecting experimental variables and evaluating possible reaction

tendencies in the metal–slag–crucible system. The Equilib module was used for equilibrium calculations. The FactPS and FToxid databases were selected to consider pure substances, oxide compounds, metallic phases, and oxide slag phases relevant to the present Fe-based smelting system. The possible equilibrium phases included gas species, metallic phases, liquid oxide slag phases, solid oxide phases, and possible intermediate oxide compounds, depending on the calculation system. The total pressure was fixed at 1 atm. The input compositions for the thermodynamic calculations were simplified based on the major components of the experimental system, including Nd, Fe-based metal, Fe_2O_3 , FeO – SiO_2 -based slag components, and crucible-related oxides such as Al_2O_3 , MgO , and ZrO_2 . Minor components in the industrial non-ferrous smelting slag were not fully included in the thermodynamic calculations unless they were directly related to each calculation case. Therefore, the calculations were not intended to reproduce the complete chemical complexity of the actual smelting experiments. It should be noted that the FactSage calculations were used only as qualitative thermodynamic guidance. The actual smelting experiments were practical high-temperature smelting experiments and were not equilibrium experiments. Therefore, the calculated equilibrium results may differ from the experimental behavior because of reaction kinetics, local oxygen potential, slag coverage, metal–slag contact, crucible–slag interaction, and limited reaction time. Accordingly, the experimental Nd distribution and recovery results were used as the main basis for evaluating Nd recovery behavior.

To evaluate the uncertainty of the Nd recovery and mass balance calculations, error propagation was applied based on the uncertainties associated with phase mass measurement and inductively coupled plasma (ICP) analysis. The amount of Nd in each phase was calculated from the measured phase mass and the Nd concentration determined by ICP analysis. The Nd amount, recovery, and loss were calculated as follows:

$$\text{Nd in phase (g)} = \text{phase mass (g)} \times \text{Nd concentration (ppm)} / 10^6$$

$$\text{Nd recovery into phase (\%)} = [\text{Nd in phase (g)} / \text{total Nd input (g)}] \times 100$$

$$\text{Overall Nd loss (\%)} = 100 - \text{Nd recovery into slag (\%)} - \text{Nd recovery into metal (\%)}$$

The relative uncertainty of the Nd amount in each phase was estimated by error propagation:

$$u(\text{Nd in phase}) / \text{Nd in phase} = [(u(m)/m)^2 + (u(C)/C)^2]^{1/2}$$

where m is the measured phase mass, C is the Nd concentration determined by ICP analysis, $u(m)$ is the uncertainty of mass measurement, and $u(C)$ is the uncertainty of ICP analysis. A relative ICP analytical uncertainty of 3% was applied for estimating the propagated uncertainty of Nd recovery. The overall Nd loss uncertainty was calculated from the propagated uncertainties of the Nd recoveries into the slag and metal phases.

In this study, X-ray fluorescence (XRF) analysis was used mainly to compare the chemical composition trends of the recovered slags, whereas the quantitative Nd mass balance and recovery values were calculated using ICP results and measured phase masses. Because the high-temperature smelting experiments were destructive and required phase separation after cooling, repeated experiments for all conditions were not performed. Therefore, the uncertainty values reported in the revised tables represent estimated analytical and measurement uncertainties rather than standard deviations from repeated smelting experiments.

The main experimental variables were the slag flux composition, slag flux addition ratio, Fe_2O_3 addition amount, slag flux type, and crucible material. First, synthetic fayalite slag flux was used as the reference slag system to determine the appropriate slag composition and addition ratio. The amount of Fe_2O_3 was then varied to evaluate its effect on Nd

oxidation and slag-phase partitioning. After the optimum conditions were identified, non-ferrous smelting slag flux was applied under the same conditions to assess its feasibility as an industrial slag flux. Finally, the effect of crucible material was examined to evaluate possible interactions among the crucible, slag, and Nd oxide during the high-temperature smelting process.

2.5.1. Selection of Slag Flux Composition

The slag flux composition is a critical factor affecting slag fluidity, metal–slag separation, and the dissolution behavior of Nd oxide in the slag phase. In this study, an FeO–SiO₂-based fayalite slag flux was selected as the reference slag system because fayalite-based slags generally have relatively low melting temperatures and suitable fluidity at high temperatures. Stoichiometric fayalite, Fe₂SiO₄, corresponds to an FeO-rich FeO–SiO₂ composition, which is favorable for forming a molten slag phase during smelting. The formation of a liquid slag phase is advantageous for improving slag fluidity, promoting metal–slag separation, and enhancing the dissolution and capture of Nd₂O₃ generated by selective oxidation of Nd.

The thermodynamic calculation was used only as qualitative guidance for selecting the reference slag composition and was not intended to quantitatively predict the actual high-temperature slag behavior in the experimental system. In the FeO–SiO₂ slag system, FeO can act as a network modifier and partially depolymerize the silicate structure, thereby improving slag fluidity and mass transfer between the metal and slag phases. However, excessive FeO content may alter the oxygen potential of the slag–metal system, whereas excessive SiO₂ content may increase slag viscosity due to the formation of a polymerized silicate network. Therefore, the fayalite-based FeO–SiO₂ composition was selected by considering liquid-phase formation, slag fluidity, and the expected dissolution behavior of Nd₂O₃ at the experimental temperature.

2.5.2. Selection of Slag Flux Addition Ratio

The slag flux addition ratio was selected by considering the need to provide sufficient slag volume for dissolving and capturing Nd oxide while avoiding unnecessary dilution of the rare-earth-bearing slag. If the slag flux addition is too low, the generated Nd₂O₃ may not be fully dissolved or captured in the slag phase and may remain in the metal phase, at the metal–slag interface, or in crucible-reacted regions. Conversely, excessive slag addition can increase the total slag volume and dilute the Nd concentration in the recovered slag, which may reduce the efficiency of subsequent rare-earth recovery from the slag. Therefore, instead of relying on thermodynamic calculation results as direct evidence, the slag flux addition ratio was treated as an experimental variable in this study. The slag flux addition ratio was set to 10, 20, and 30 wt% based on the total mass of the experimental charge. This range was selected to evaluate the effect of increasing slag volume on Nd transfer from the Fe-based metal phase to the slag phase and to identify a suitable slag flux addition condition for Nd recovery.

2.5.3. Selection of Oxidizing Agent

Fe₂O₃ was selected as the oxidizing agent to promote the selective oxidation of Nd in the molten metal phase. The thermodynamic calculation results indicate that the oxidation of Nd to Nd₂O₃ is thermodynamically favorable over the investigated temperature range, confirming the strong oxygen affinity of Nd. In the proposed process, the rotor-derived steel and Nd–Fe–B magnet fractions were first melted, and the oxidizing agent was then added to induce the preferential oxidation of Nd in the molten metal phase. Fe₂O₃ supplies oxygen at high temperature and can oxidize Nd according to the following reaction:



This reaction allows Nd dissolved in the molten metal phase to be converted into Nd_2O_3 . After sufficient oxidation, slag flux was added to dissolve and capture the generated Nd_2O_3 in the slag phase. Therefore, in this process, the main role of the slag flux is not to directly oxidize Nd, but to stabilize and collect the Nd oxide formed by Fe_2O_3 -assisted oxidation. In other words, the Nd oxidation step and the slag capture step were designed as sequential process stages. The selection of Fe_2O_3 was based on its compatibility with the Fe-based smelting system rather than on thermodynamic calculation alone. The reduced product of Fe_2O_3 is Fe, which can be incorporated into the Fe-rich metallic phase without introducing additional foreign metallic impurities. In contrast, other oxides such as MnO_2 , NiO , and CuO may introduce Mn, Ni, or Cu into the metallic phase after reduction, which can complicate process control and product separation. Therefore, Fe_2O_3 was selected as a suitable oxidizing agent for promoting Nd oxidation while maintaining compatibility with the Fe-based metal phase.

2.5.4. Selection of Fe_2O_3 Addition Amount

The Fe_2O_3 addition amount was selected as an important experimental variable because it directly affects the oxygen potential of the metal–slag system and the extent of Nd oxidation. Fe_2O_3 was used to promote the selective oxidation of Nd dissolved in the Fe-based molten metal phase. Insufficient Fe_2O_3 addition may limit Nd oxidation and reduce the transfer of Nd into the slag phase. In contrast, excessive Fe_2O_3 addition may alter the slag composition, increase gas formation through side reactions such as carbon oxidation, and affect the stability of the smelting process. In the original manuscript, thermodynamic calculations were used to support the selection of the Fe_2O_3 addition range. However, because the calculation conditions did not fully reproduce the actual experimental smelting system, the revised manuscript does not use these calculations as direct quantitative evidence. Instead, the Fe_2O_3 addition amount was treated as an experimental variable, and its effect on Nd recovery was evaluated primarily based on the experimental metal–slag separation results. Therefore, the Fe_2O_3 addition amount was varied from 0 to 4 wt% based on the total mass of the experimental charge. This range was selected to include insufficient, suitable, and excessive oxidizing-agent conditions and to determine the experimentally appropriate Fe_2O_3 addition amount for Nd oxidation and slag-phase partitioning. The experimental results were then used to identify the optimum Fe_2O_3 addition condition for maximizing Nd transfer into the slag phase while maintaining stable process behavior.

2.5.5. Selection of Reaction Temperature

The reaction temperature was selected to ensure complete melting of the metallic charge and sufficient reactivity for selective Nd oxidation and subsequent metal–slag separation. In the proposed process, the rotor-derived steel and Nd–Fe–B magnet fractions were first melted together, followed by Fe_2O_3 addition for Nd oxidation and slag flux addition for Nd oxide capture. Therefore, achieving a fully molten and homogeneous metal phase before oxidation and slagging was essential. The experimental charge consisted mainly of steel with a small amount of Nd–Fe–B magnet. Because the steel fraction was dominant, a temperature higher than the melting range of the Fe-based metallic charge was required to ensure complete melting and stable metal–slag reaction. Although thermodynamic phase information can provide a useful reference for estimating the melting behavior of the Fe-based charge, the actual experimental temperature was selected by considering practical smelting conditions, melt fluidity, reaction kinetics, and stable metal–slag separation. At temperatures close to the melting range, incomplete melting, insufficient homogenization, or partial solid retention may occur, which can hinder the contact between Nd in the molten metal and the oxidizing agent. In addition, relatively low temperatures may increase melt viscosity and reduce diffusion rates, thereby limiting the kinetics of Nd oxidation and metal–slag reactions. Therefore, the reaction temperature was set to 1600 °C to ensure complete melting, sufficient fluidity, and enhanced mass transfer in the molten bath. This temperature was used as a practical smelting condition for promoting Nd oxidation by Fe_2O_3 and facilitating the subsequent dissolution and capture of Nd_2O_3 in the slag phase.

2.5.6. Application Design of Non-Ferrous Smelting Slag Flux

In this study, non-ferrous smelting slag flux was applied as an alternative to the synthesized fayalite slag flux to evaluate the practical applicability of the proposed pyrometallurgical Nd recovery process. The synthesized fayalite slag flux was first used as a reference slag system to determine the suitable slag-forming conditions. Subsequently, non-ferrous smelting slag flux generated from an actual industrial process was applied under the selected conditions to examine whether it could provide a similar function in Nd oxide dissolution and slag-phase capture. The phase characteristics of the synthesized fayalite slag flux and non-ferrous smelting slag flux were compared by X-ray diffraction (XRD), as shown in Figure 2. The XRD results confirmed that Fe_2SiO_4 , corresponding to the fayalite phase, was detected in both slag fluxes. This indicates that the non-ferrous smelting slag flux has room-temperature phase characteristics broadly similar to those of the synthesized fayalite slag flux. It should be noted that the XRD results do not directly represent the properties of the liquid slag at high temperature, such as viscosity, oxygen potential, or Nd_2O_3 solubility. Therefore, in this study, XRD analysis was used only to compare the initial phase characteristics of the slag fluxes. The high-temperature applicability of the non-ferrous smelting slag flux was evaluated mainly based on the experimental Nd recovery behavior and slag–metal separation results.

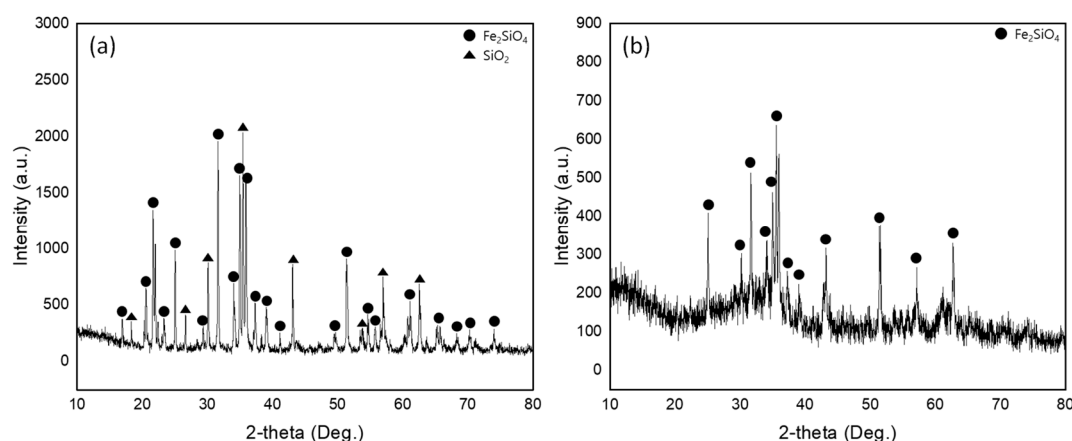


Figure 2. XRD patterns of slag fluxes used in this study: (a) synthesized fayalite slag flux and (b) non-ferrous smelting slag flux.

The chemical compositions of the synthesized fayalite-based slag flux and non-ferrous smelting slag flux were compared by X-ray fluorescence (XRF), as summarized in Table 1. The non-ferrous smelting slag flux mainly consisted of total Fe, expressed as Fe₂O₃ equivalent, and SiO₂, with contents of 54.44 wt% and 30.69 wt%, respectively. This composition is broadly comparable to that of the synthesized fayalite-based slag flux, which contained 55.45 wt% total Fe expressed as Fe₂O₃ equivalent and 41.49 wt% SiO₂. It should be noted that the Fe₂O₃ values reported in Table 1 represent total iron expressed as Fe₂O₃ equivalent by XRF analysis and do not indicate that all iron exists as Fe₂O₃ in the slag. The actual FeO/Fe₂O₃ ratio was not directly determined in this study. However, unlike the synthesized fayalite-based slag flux, the non-ferrous smelting slag flux contained additional oxides such as Al₂O₃, MgO, CaO, CuO, ZnO, and K₂O, reflecting its origin as an industrial by-product.

Table 1. Chemical compositions of synthesized fayalite slag flux and non-ferrous smelting slag flux determined by XRF.

Material	Total Fe as Fe ₂ O ₃ Equivalent (wt%)	SiO ₂ (wt%)	Al ₂ O ₃ (wt%)	MgO (wt%)	CaO (wt%)	CuO (wt%)	ZnO (wt%)	K ₂ O (wt%)	Others (wt%)
Non-ferrous smelting slag flux	54.44	30.69	4.82	1.93	1.85	1.41	1.16	1.05	2.65
Synthesized fayalite slag flux	55.45	41.49	0.8	1.51	—	—	—	—	0.66

Note: The Fe content is reported as total Fe expressed as Fe₂O₃ equivalent by XRF analysis. The actual FeO/Fe₂O₃ ratio was not directly determined in this study.

These additional oxide components may influence the physicochemical properties of the slag. Al₂O₃ and MgO can act as network-forming or intermediate oxides and may increase slag viscosity by strengthening the slag structure. In contrast, basic oxides such as CaO can partially depolymerize the SiO₂-based network structure, thereby affecting slag fluidity and oxide dissolution behavior. Transition metal oxides such as CuO and ZnO may also participate in oxidation–reduction reactions, which can influence the oxygen potential of the slag–metal system and the distribution behavior of Nd. Although synthesized fayalite slag flux is useful for clarifying thermodynamic behavior and reaction mechanisms because of its relatively simple FeO–SiO₂-based composition, actual industrial slags contain multiple oxide components. Therefore, applying non-ferrous smelting slag flux is important for evaluating the practical feasibility of the proposed process. In this study, the comparison between synthesized fayalite slag flux and non-ferrous smelting

slag flux was used to assess whether an industrial slag by-product could be used as a functional flux for Nd recovery from end-of-life EV motor materials.

3. Results and Discussion

3.1. Effect of Fayalite Slag Flux Addition on Nd Recovery Behavior

The effect of fayalite slag flux addition on Nd recovery behavior was investigated by fixing the Fe_2O_3 addition at 2 wt% based on the mass of the rotor-derived charge and varying the fayalite slag flux addition ratio from 10 to 30 wt%. The experiments were conducted under an air atmosphere to evaluate Nd oxidation and metal–slag separation behavior under practically relevant oxidizing conditions. Fe_2O_3 was selected as the oxidizing agent based on the thermodynamic analysis described in Section 2.5.4, which indicated that Nd can be effectively oxidized to Nd_2O_3 at relatively low Fe_2O_3 addition levels. The experimental charge consisted of steel and Nd–Fe–B magnet fractions, corresponding to the rotor-derived composition. The steel and magnet ratios were fixed at 94.32 and 5.68 wt%, respectively, and the fayalite slag flux was added at 10, 20, and 30 wt% relative to the total mass of the charge. The experimental conditions are summarized in Table 2. Alumina crucibles were used in this series of experiments.

Table 2. Experimental conditions with different fayalite slag flux addition ratios.

Material/Condition	Value	Weight Based on 100 g Rotor-Derived Charge
Steel	94.32 wt%	94.32 g
Nd–Fe–B magnet	5.68 wt%	5.68 g
Estimated Nd in magnet fraction	1.70 wt%	1.70 g
Total rotor-derived charge	100 wt%	100.00 g
Fe_2O_3 addition	2 wt% of total charge	2.00 g
Fayalite-based slag flux addition	10, 20, and 30 wt% of total charge	10.00, 20.00, and 30.00 g
Atmosphere	Air	–
Crucible type	Alumina crucible	–

Note: The listed weights are calculated based on 100 g of rotor-derived charge. The Fe_2O_3 and slag flux additions were calculated relative to the mass of the rotor-derived charge.

After smelting, the slag phase was separated from the metal phase, crushed, and analyzed by X-ray fluorescence (XRF) to evaluate the change in slag composition with increasing fayalite slag flux addition. The chemical compositions of the slags are summarized in Table 3. The slags mainly consisted of SiO_2 , total Fe expressed as Fe_2O_3 equivalent, Al_2O_3 , and Nd_2O_3 , indicating that the oxidized Nd was transferred into the slag phase during the smelting process. It should be noted that the Fe_2O_3 values in Table 3 represent total Fe expressed as Fe_2O_3 equivalent by XRF analysis and do not indicate the actual $\text{FeO}/\text{Fe}_2\text{O}_3$ ratio in the slag. The Nd_2O_3 content in the slag was 6.58, 4.12, and 6.58 wt% at fayalite slag flux addition ratios of 10, 20, and 30 wt%, respectively. Although the Nd_2O_3 concentration in the slag did not increase monotonically with increasing slag addition, the total amount of slag recovered increased significantly. Therefore, the Nd_2O_3 concentration alone is not sufficient to evaluate the Nd recovery behavior; the total mass of slag and the Nd concentration in the slag must be considered together.

Table 3. Chemical composition of slag with different fayalite slag flux addition ratios.

Component	10 wt%	20 wt%	30 wt%
SiO_2	47.52	48.24	48.54

Fe ₂ O ₃	24.08	20.02	15.87
Al ₂ O ₃	19.07	23.45	25.12
Nd ₂ O ₃	5.09	4.12	6.58
MnO	3.02	2.89	2.91
Cr ₂ O ₃	0.35	0.39	0.19
CaO	0.11	0.07	–
TiO ₂	0.53	0.41	0.29
V ₂ O ₅	0.09	0.17	0.21
Dy ₂ O ₃	0.14	0.24	0.29
Total	100	100	100

Note: The Fe content is reported as total Fe expressed as Fe₂O₃ equivalent by XRF analysis. Therefore, the values do not represent the actual FeO/Fe₂O₃ ratio in the recovered slag.

The Al₂O₃ content was relatively high in all slag samples, ranging from 23.45 to 25.12 wt%, which is attributed to the partial dissolution or incorporation of the alumina crucible into the slag under high-temperature smelting conditions. This crucible-derived Al₂O₃ may have modified the slag structure and affected the dissolution behavior of Nd-containing oxide species. The lower total Fe content expressed as Fe₂O₃ equivalent in the recovered slag compared with the initial fayalite-based flux can be attributed to changes in slag composition during smelting. In particular, the incorporation of crucible-derived Al₂O₃ and newly formed Nd₂O₃ diluted the relative concentration of iron oxide in the recovered slag. In addition, part of the iron oxide may have participated in oxidation–reduction reactions and been reduced to Fe or FeO under local metal–slag reaction conditions, with reduced Fe being incorporated into the Fe-rich metallic phase. Therefore, the recovered slag composition reflects the final reaction products after smelting rather than the original flux composition alone.

To quantitatively evaluate Nd distribution, the separated slag and metal phases were analyzed by ICP, and the Nd recovery was calculated based on the measured mass and Nd concentration of each phase using the recovery equation described in Section 2.5. The results are summarized in Table 4. As the fayalite slag flux addition ratio increased, the Nd recovery into the slag phase increased markedly. At 10 wt% slag flux addition, 1.24 g of Nd was recovered in the slag phase, corresponding to a recovery rate of 40.12%. When the slag flux addition ratio was increased to 20 wt%, the amount of Nd in the slag increased to 2.51 g, and the recovery rate improved to 65.03%. The highest Nd recovery of 80.92% was obtained at 30 wt% slag flux addition, with 2.63 g of Nd recovered in the slag phase.

Table 4. Effect of fayalite slag flux addition on Nd distribution and recovery.

Fayalite Slag Flux Addition	10 wt%	20 wt%	30 wt%
Total Nd			
Amount of Nd in simulated material (g)	3.09	3.86	3.25
Slag phase			
Slag after smelting (g)	19.05	41.08	53.44
Nd in slag by ICP (ppm)	64,989	61,115	49,135
Nd in slag (g)	1.24	2.51	2.63
Nd recovery into slag (%)	40.12 ± 1.20	65.03 ± 1.95	80.92 ± 2.43
Metal phase			
Metal after smelting (g)	175.55	221.29	187.44
Nd in metal by ICP (ppm)	103	28	13
Nd in metal (g)	0.02	0.01	0.002
Nd recovery into metal (%)	0.65 ± 0.02	0.26 ± 0.01	0.07 ± 0.01

Overall Nd loss during smelting (g)	1.83	1.34	0.62
Overall Nd loss during smelting (%)	59.23 ± 1.20	34.71 ± 1.95	19.01 ± 2.43

Note: The overall Nd loss was calculated as the difference between the total Nd input and the Nd recovered in the slag and metal phases. The loss term includes unmeasured Nd associated with crucible interaction, adhered slag or metal, incomplete phase separation, sampling loss, and other process-related losses. The uncertainty values were estimated by error propagation using phase mass measurements and a relative ICP uncertainty of 3%.

The increase in Nd recovery with increasing slag flux addition can be attributed to the enhanced capacity of the slag phase to dissolve and retain Nd-containing oxides. When the slag flux amount was insufficient, the generated Nd oxide could not be fully incorporated into the slag phase and may have remained at the metal–slag interface or been lost through incomplete phase separation. In contrast, increasing the slag flux addition provided a larger slag volume and higher oxide dissolution capacity, thereby promoting the transfer of Nd from the metal phase to the slag phase. The Nd concentration remaining in the metal phase decreased with increasing slag flux addition. The Nd concentration in the metal was 103 ppm at 10 wt% slag flux addition, decreased to 28 ppm at 20 wt%, and further decreased to 13 ppm at 30 wt%. This result confirms that the transfer of Nd from the metal phase to the slag phase was enhanced as the slag flux addition ratio increased. Therefore, under the present experimental conditions, 30 wt% fayalite slag flux was identified as the most effective condition for Nd recovery. XRD analysis was conducted on the slag recovered after smelting to identify the crystalline phases containing Nd. As shown in Figure 3, the XRD pattern did not show distinct crystalline peaks corresponding to Nd_2O_3 or other Nd-containing crystalline phases. Instead, a broad amorphous-like pattern was observed. This suggests that the recovered slag mainly formed a glassy or poorly crystallized structure after high-temperature melting and cooling.

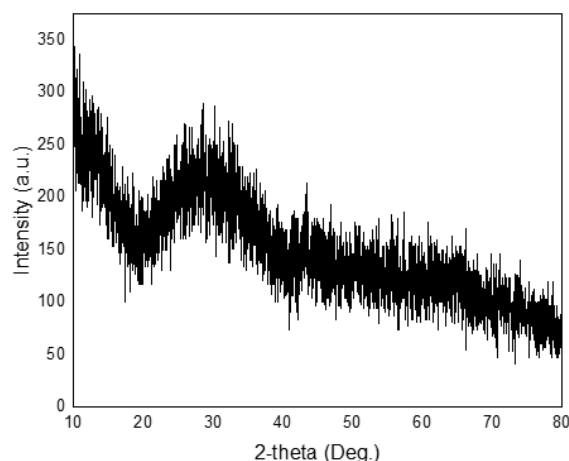


Figure 3. XRD pattern of slag recovered after smelting.

The absence of clear Nd_2O_3 peaks does not necessarily indicate that Nd was not incorporated into the slag. The XRF and ICP results confirmed that a significant amount of Nd was present in the slag phase. Therefore, Nd is likely to exist as dissolved Nd-containing oxide species within the silicate-based slag matrix rather than as a well-crystallized Nd_2O_3 phase. Considering the high SiO_2 content of the slag and the incorporation of Al_2O_3 from the alumina crucible, the slag structure is expected to favor glass formation during cooling. Under such conditions, Nd may be incorporated into the amorphous silicate network or form finely dispersed Nd-containing silicate species that are difficult to detect by conventional XRD. Previous thermodynamic assessments of the Nd_2O_3 – CaO – SiO_2 system

have shown that Nd-containing silicate phases can be stabilized in silicate-based oxide systems [21]. Based on this thermodynamic tendency, together with the amorphous XRD pattern and the chemical analysis results, the recovered Nd in this study is considered to be stabilized mainly within the silicate-based slag matrix, possibly as dissolved Nd oxide or Nd-silicate species. Therefore, the slag flux addition ratio plays a critical role not only in providing sufficient slag volume but also in stabilizing oxidized Nd within the slag phase.

3.2. Effect of Fe_2O_3 Addition on Nd Recovery Behavior

The effect of Fe_2O_3 addition on Nd recovery behavior was investigated to evaluate the role of the oxidizing agent in promoting the selective oxidation of Nd and its subsequent transfer into the slag phase. Based on the results of the fayalite slag flux addition experiments, the slag flux addition ratio was fixed at 30 wt%, which showed the highest Nd recovery. The Fe_2O_3 addition amount was then varied from 0 to 4 wt% based on the mass of the rotor-derived charge. The experiments were conducted under an air atmosphere using alumina crucibles. The experimental conditions are summarized in Table 5.

Table 5. Experimental conditions as a function of Fe_2O_3 addition.

Material/Condition	Value	Weight Based on 100 g Rotor-Derived Charge
Steel	94.32 wt%	94.32 g
Nd–Fe–B magnet	5.68 wt%	5.68 g
Estimated Nd in magnet fraction	1.70 wt%	1.70 g
Total rotor-derived charge	100 wt%	100.00 g
Fe_2O_3 addition	0, 1, 2, 3, and 4 wt% of total charge	0.00, 1.00, 2.00, 3.00, and 4.00 g
Fayalite-based slag flux addition	30 wt% of total charge	30.00 g
Atmosphere	Air	–
Crucible type	Alumina crucible	–

Note: The listed weights are calculated based on 100 g of rotor-derived charge. The Fe_2O_3 and slag flux additions were calculated relative to the mass of the rotor-derived charge.

It should be noted that the present experiments were practical smelting experiments conducted in an air atmosphere and were not equilibrium experiments between the molten alloy and air. During smelting, the molten Fe-based alloy was in contact with the slag phase, and the slag layer partially covered the molten metal surface. Therefore, direct oxidation of the entire molten alloy by atmospheric oxygen was kinetically limited, although surface oxidation and slag oxidation reactions could occur. In this process, the oxidation of Nd was intentionally promoted by Fe_2O_3 addition, and the generated Nd oxide was subsequently dissolved and captured in the slag phase. Thus, the local oxygen potential at the metal–slag interface was affected by Fe_2O_3 addition, slag composition, and metal–slag reactions, rather than by atmospheric oxygen alone.

Fe_2O_3 was added after melting the steel and Nd–Fe–B magnet fractions to promote the oxidation of Nd in the molten metal phase. Although the thermodynamic analysis indicated that Nd oxidation can occur at relatively low Fe_2O_3 addition levels, the actual smelting process may be affected by reaction kinetics, mixing conditions, local oxygen availability, slag coverage, and metal–slag contact. Therefore, the Fe_2O_3 addition range was experimentally evaluated to determine the appropriate amount required for effective Nd oxidation and slag-phase recovery. Excessive Fe_2O_3 addition was also examined because it may promote side reactions such as carbon oxidation and gas formation, which can affect process stability.

The chemical compositions of the slags obtained after smelting are summarized in Table 6. The Fe content in Table 6 is reported as total Fe expressed as Fe_2O_3 equivalent by XRF analysis and does not represent the actual $\text{FeO}/\text{Fe}_2\text{O}_3$ ratio in the slag. The actual $\text{FeO}/\text{Fe}_2\text{O}_3$ ratio was not directly determined in this study. Therefore, the Fe_2O_3 -equivalent values should be interpreted as total Fe contents expressed in oxide form rather than as direct evidence of the oxidation state of Fe in the molten slag. The Nd_2O_3 content in the slag increased with Fe_2O_3 addition. In the absence of Fe_2O_3 , the Nd_2O_3 content in the slag was 2.31 wt%, indicating that a limited amount of Nd was oxidized and transferred to the slag phase even under the air atmosphere. When 1 wt% Fe_2O_3 was added, the Nd_2O_3 content increased to 3.38 wt%. A further increase in Fe_2O_3 addition to 2 wt% resulted in a significant increase in Nd_2O_3 content to 6.58 wt%, suggesting that Nd oxidation was strongly promoted under this condition. At 3 and 4 wt% Fe_2O_3 addition, the Nd_2O_3 contents were 6.14 and 7.29 wt%, respectively, indicating that the Nd_2O_3 concentration remained at a relatively high level once sufficient oxidizing agent was supplied. The total Fe content expressed as Fe_2O_3 equivalent in the recovered slags was lower than that in the initial slag flux because the slag composition changed during smelting. The incorporation of newly formed Nd_2O_3 and crucible-derived Al_2O_3 diluted the relative concentration of iron oxide in the recovered slag. In addition, part of the iron oxide may have participated in oxidation–reduction reactions during Nd oxidation and carbon oxidation, and reduced Fe could be incorporated into the Fe-rich metallic phase. Therefore, the final Fe content in the recovered slag reflects the combined effects of slag dilution, crucible interaction, and metal–slag redox reactions, rather than the original flux composition alone.

Table 6. Chemical composition of slag with varying Fe_2O_3 addition.

Component	0 wt%	1 wt%	2 wt%	3 wt%	4 wt%
SiO_2	53.97	52.17	48.54	44.31	46.37
Total Fe as Fe_2O_3 equivalent	13.16	13.97	15.87	19.68	17.58
Al_2O_3	27.15	26.33	25.12	26.16	25.89
Nd_2O_3	2.31	3.38	6.58	6.14	7.29
MnO	2.78	3.21	2.91	3.05	2.31
Cr_2O_3	0.13	0.25	0.19	0.13	0.11
CaO	–	0.07	–	0.02	–
TiO_2	0.37	0.33	0.29	0.3	0.31
V_2O_5	0.13	0.15	0.21	0.21	0.14
Dy_2O_3	–	0.14	0.29	–	–
Total	100	100	100	100	100

Note: The Fe content is reported as total Fe expressed as Fe_2O_3 equivalent by XRF analysis. Therefore, the values do not represent the actual $\text{FeO}/\text{Fe}_2\text{O}_3$ ratio in the slag.

The SiO_2 content decreased overall as the Fe_2O_3 addition increased, from 53.97 wt% at 0 wt% Fe_2O_3 to 44.31 wt% at 3 wt% Fe_2O_3 . This decrease is mainly attributed to the relative increase in iron oxide and Nd_2O_3 contents in the slag. The total Fe content expressed as Fe_2O_3 equivalent increased with Fe_2O_3 addition, although the increase was not strictly linear. This suggests that part of the added Fe_2O_3 participated in oxidation reactions with Nd and other elements, while the remaining fraction was retained in the slag phase as iron oxide species. Because the $\text{FeO}/\text{Fe}_2\text{O}_3$ ratio is strongly dependent on oxygen potential, the redox state of iron oxide in the molten slag may influence slag physicochemical properties such as oxygen potential, viscosity, and oxide dissolution behavior. However, direct $\text{Fe}^{2+}/\text{Fe}^{3+}$ analysis was not performed in this study. Therefore, the effect of slag redox state on Nd recovery should be investigated in future work. The Al_2O_3 content

remained relatively high, at approximately 25–27 wt%, which is attributed to interaction between the slag and the alumina crucible during high-temperature smelting. These results indicate that Fe_2O_3 addition not only promotes Nd oxidation but also modifies the slag composition and potentially affects slag structure and fluidity. The distribution of Nd between the slag and metal phases was quantitatively evaluated by ICP analysis, and the results are summarized in Table 7. The Nd recovery into the slag phase increased sharply with Fe_2O_3 addition. Without Fe_2O_3 addition, the Nd recovery into the slag phase was 36.89%. This indicates that some Nd oxidation occurred under the air atmosphere and in the presence of fayalite slag flux; however, the oxidation and transfer of Nd were not sufficient. When 1 wt% Fe_2O_3 was added, the Nd recovery increased slightly to 41.97%. A marked improvement was observed at 2 wt% Fe_2O_3 addition, where the amount of Nd recovered in the slag reached 2.63 g and the recovery rate increased to 80.92%.

Table 7. Effect of Fe_2O_3 addition on Nd distribution and recovery.

Fe_2O_3 Addition	0 wt%	1 wt%	2 wt%	3 wt%	4 wt%
Total Nd					
Amount of Nd in simulated material (g)	2.98	3.11	3.25	3.51	3.03
Slag phase					
Slag after smelting (g)	49.14	52.39	53.44	60.31	51.08
Nd in slag by ICP (ppm)	23,185	31,003	49,135	46,250	47,058
Nd in slag (g)	1.14	1.62	2.63	2.79	2.4
Nd recovery into slag (%)	36.89 ± 1.11	41.97 ± 1.26	80.92 ± 2.43	79.49 ± 2.38	79.21 ± 2.38
Metal phase					
Metal after smelting (g)	170.51	178.58	187.44	210.58	179.21
Nd in metal by ICP (ppm)	46	31	13	10	11
Nd in metal (g)	0.01	0.01	0.002	0.002	0.002
Nd recovery into metal (%)	0.33 ± 0.01	0.32 ± 0.01	0.07 ± 0.01	0.06 ± 0.01	0.07 ± 0.01
Overall Nd mass balance					
Overall Nd loss during smelting (g)	1.83	1.48	0.62	0.72	0.63
Overall Nd loss during smelting (%)	62.78 ± 1.11	57.71 ± 1.26	19.01 ± 2.43	20.45 ± 2.38	20.72 ± 2.38

Note: The overall Nd loss was calculated as the difference between the total Nd input and the Nd recovered in the slag and metal phases. The loss term includes unmeasured Nd associated with crucible interaction, adhered slag or metal, incomplete phase separation, sampling loss, and other process-related losses. The uncertainty values were estimated by error propagation using phase mass measurements and a relative ICP uncertainty of 3%.

When the Fe_2O_3 addition was further increased to 3 and 4 wt%, the Nd recovery into the slag phase was 79.49 and 79.21%, respectively. These values were similar to that obtained at 2 wt% Fe_2O_3 , indicating that additional Fe_2O_3 above 2 wt% did not significantly improve Nd recovery. This suggests that the oxidation of Nd was already sufficiently promoted at 2 wt% Fe_2O_3 under the present experimental conditions. Further addition of Fe_2O_3 may therefore be unnecessary and may instead increase the possibility of side reactions, gas generation, and slag composition changes. The Nd concentration remaining in the metal phase decreased as Fe_2O_3 was added. The Nd content in the metal phase was 46 ppm without Fe_2O_3 addition and decreased to 31 ppm at 1 wt% Fe_2O_3 . At Fe_2O_3 addition levels of 2 wt% or higher, the Nd content in the metal phase decreased to approximately 10–13 ppm. This indicates that most of the Nd was removed from the metal phase and transferred to the slag phase when sufficient Fe_2O_3 was supplied. The residual Nd amount in the metal phase was extremely small and had a negligible effect on the overall recovery calculation. The recovery of an Fe-rich metallic phase after smelting also indicates that complete oxidation of the alloy did not occur under the present experimental conditions,

even though the experiments were conducted in air. Overall, Fe_2O_3 addition strongly affected Nd oxidation and slag-phase recovery. The Nd recovery increased sharply up to 2 wt% Fe_2O_3 addition, whereas further increases in Fe_2O_3 addition did not lead to a meaningful improvement in recovery. Therefore, 2 wt% Fe_2O_3 was identified as the optimum oxidizing agent addition amount under the present conditions. This condition provided sufficient Nd oxidation and high Nd transfer into the slag phase while minimizing unnecessary oxidizer consumption and possible side reactions. However, because the $\text{FeO}/\text{Fe}_2\text{O}_3$ ratio and oxygen potential of the molten slag were not directly measured, future work should include direct $\text{Fe}^{2+}/\text{Fe}^{3+}$ analysis and oxygen potential control to more accurately evaluate the relationship between slag redox state, slag physicochemical properties, and Nd recovery behavior.

3.3. Application of Non-Ferrous Smelting Slag Flux

Based on the optimum conditions obtained from the experiments using synthesized fayalite slag flux, additional experiments were conducted using non-ferrous smelting slag flux to evaluate the practical applicability of the proposed process. The non-ferrous smelting slag flux was generated from an actual industrial non-ferrous smelting process and contained Fe–Si–O-based components similar to those of the synthesized fayalite slag flux. Therefore, it was expected to act as a functional slag-forming material for dissolving and capturing Nd-containing oxide species. To directly compare the effect of slag flux type, the experimental conditions were fixed at the optimum conditions identified in the previous sections. The slag flux addition ratio was fixed at 30 wt% of the total charge, and the Fe_2O_3 addition was fixed at 2 wt% of the total charge. The experiments were conducted under an air atmosphere using alumina crucibles. Under these identical conditions, synthesized fayalite slag flux and non-ferrous smelting slag flux were applied separately. The experimental conditions are summarized in Table 8.

Table 8. Experimental conditions for comparing different slag flux types.

Material/Condition	Value	Weight Based on 100 g Rotor-Derived Charge
Steel	94.32 wt%	94.32 g
Nd–Fe–B magnet	5.68 wt%	5.68 g
Estimated Nd in magnet fraction	1.70 wt%	1.70 g
Total rotor-derived charge	100 wt%	100.00 g
Fe_2O_3 addition	2 wt% of total charge	2.00 g
Slag flux addition	30 wt% of total charge	30.00 g
Slag flux type	Fayalite-based slag flux or non-ferrous smelting slag flux	30.00 g
Atmosphere	Air	–
Crucible type	Alumina crucible	–

Note: The listed weights are calculated based on 100 g of rotor-derived charge. For the slag flux comparison experiments, either 30.00 g of fayalite-based slag flux or 30.00 g of non-ferrous smelting slag flux was added.

The chemical compositions of the slags obtained after smelting with different slag flux types are summarized in Table 9. Both slags mainly consisted of SiO_2 , total Fe expressed as Fe_2O_3 equivalent, Al_2O_3 , and Nd_2O_3 . It should be noted that the Fe content in Table 9 is reported as total Fe expressed as Fe_2O_3 equivalent by XRF analysis and does not represent the actual $\text{FeO}/\text{Fe}_2\text{O}_3$ ratio in the recovered slag. The SiO_2 contents were 48.54 wt% for the fayalite-based slag flux condition and 50.50 wt% for the non-ferrous smelting

slag flux condition, indicating that both recovered slags retained a silicate-based composition after smelting.

Table 9. Chemical composition of recovered slags obtained using fayalite slag flux and non-ferrous smelting slag flux.

Component	Fayalite Slag Flux	Non-Ferrous Smelting Slag Flux
SiO ₂	48.54	50.5
Total Fe as Fe ₂ O ₃ equivalent	15.87	11.99
Al ₂ O ₃	25.12	25.03
Nd ₂ O ₃	6.58	5.92
MnO	2.91	0.97
K ₂ O	–	1
Na ₂ O	–	0.54
Cr ₂ O ₃	0.19	0.26
MgO	–	1.27
CaO	–	1.85
TiO ₂	0.29	0.51
V ₂ O ₅	0.21	–
Dy ₂ O ₃	0.29	–
ZnO	–	0.16
Total	100	100

Note: The Fe content is reported as total Fe expressed as Fe₂O₃ equivalent by XRF analysis. Therefore, the values do not represent the actual FeO/Fe₂O₃ ratio in the recovered slag.

The total Fe content expressed as Fe₂O₃ equivalent was slightly lower in the non-ferrous smelting slag flux condition than in the fayalite-based slag flux condition, whereas the Al₂O₃ content was similar in both conditions at approximately 25 wt%. This relatively high Al₂O₃ content is attributed to interaction between the slag and the alumina crucible during high-temperature smelting. The lower Fe content in the recovered slags compared with the initial fluxes can be attributed to compositional changes during smelting. In particular, the incorporation of newly formed Nd₂O₃ and crucible-derived Al₂O₃ diluted the relative concentration of iron oxide in the slag. In addition, part of the iron oxide may have participated in metal–slag redox reactions and been reduced to Fe or FeO, with reduced Fe being incorporated into the Fe-rich metallic phase. Therefore, the recovered slag composition reflects the final reaction products after smelting rather than the original flux composition alone.

The Nd₂O₃ content in the recovered slag was 6.58 wt% for the fayalite slag flux condition and 5.92 wt% for the non-ferrous smelting slag flux condition. This indicates that Nd was successfully oxidized and incorporated into the slag phase in both cases. Although the non-ferrous smelting slag flux showed a slightly lower Nd₂O₃ concentration, the difference was not large, suggesting that it can provide a slag environment capable of capturing Nd-containing oxide species. Unlike the synthesized fayalite slag flux, the non-ferrous smelting slag flux contained additional oxides such as MgO, CaO, K₂O, Na₂O, and ZnO. These components reflect the multi-component nature of industrial slag by-products. MgO and Al₂O₃ may increase slag viscosity by strengthening the slag structure, whereas CaO, K₂O, and Na₂O can modify the silicate network and affect slag fluidity. Therefore, the non-ferrous smelting slag flux may exhibit more complex physicochemical behavior than the synthesized fayalite slag flux. Nevertheless, the similar SiO₂–Fe₂O₃–Al₂O₃-based composition and the presence of Nd₂O₃ in the slag indicate that non-ferrous smelting slag flux can function as an alternative slag-forming material for Nd recovery. The Nd distribution and recovery behavior obtained using the two slag flux types are

summarized in Table 10. The initial amount of Nd in the simulated material was 3.25 g for the fayalite slag flux condition and 4.21 g for the non-ferrous smelting slag flux condition. This difference is attributed to the difference in the magnet mass used in each experiment. Therefore, the Nd recovery rate was used as the main parameter for comparing the two slag flux types. It should be noted that the two experiments were not conducted at an identical melting scale. Therefore, the absolute amounts of recovered metal and slag should not be directly compared. Instead, the comparison was made mainly on the basis of the normalized Nd recovery rate. The purpose of this experiment was to evaluate the feasibility of using industrial non-ferrous smelting slag flux as an alternative slag-forming material under the optimized process conditions identified from the fayalite slag flux experiments. Nevertheless, the difference in melting scale may have affected heat transfer, slag–metal contact, mixing, and phase separation behavior. Therefore, this comparison should be interpreted as a recovery-based feasibility comparison rather than a strictly equivalent-scale comparison.

Table 10. Effect of slag flux type on Nd distribution and recovery.

Slag Flux Type	Fayalite Slag Flux	Non-Ferrous Smelting Slag Flux
Total Nd		
Amount of Nd in simulated material (g)	3.25	4.21
Slag phase		
Slag after smelting (g)	53.44	86.33
Nd in slag by ICP (ppm)	49,135	35,691
Nd in slag (g)	2.63	3.08
Nd recovery into slag (%)	80.92 ± 2.43	73.16 ± 2.19
Metal phase		
Metal after smelting (g)	187.44	256.01
Nd in metal by ICP (ppm)	13	11
Nd in metal (g)	0.002	0.003
Nd recovery into metal (%)	0.07 ± 0.01	0.07 ± 0.01
Overall Nd mass balance		
Overall Nd loss during smelting (g)	0.62	1.13
Overall Nd loss during smelting (%)	19.01 ± 2.43	26.77 ± 2.19

Note: The overall Nd loss was calculated as the difference between the total Nd input and the Nd recovered in the slag and metal phases. The loss term includes unmeasured Nd associated with crucible interaction, adhered slag or metal, incomplete phase separation, sampling loss, and other process-related losses. The uncertainty values were estimated by error propagation using phase mass measurements and a relative ICP uncertainty of 3%.

The amount of Nd recovered in the slag phase was 2.63 g for the fayalite slag flux condition and 3.08 g for the non-ferrous smelting slag flux condition. The corresponding Nd recovery rates were 80.92% and 73.16%, respectively. Although the recovery rate obtained using non-ferrous smelting slag flux was lower than that obtained using synthesized fayalite slag flux, the difference was moderate, and the overall Nd recovery behavior was comparable. The lower Nd concentration in the non-ferrous smelting slag flux condition, 35,691 ppm, compared with 49,135 ppm for the fayalite slag flux condition, can be partly attributed to the larger amount of recovered slag. The mass of recovered slag was 86.33 g for the non-ferrous smelting slag flux condition, whereas it was 53.44 g for the fayalite slag flux condition. Therefore, even though the Nd concentration in the non-ferrous smelting slag was lower, the total amount of Nd recovered in the slag phase was slightly higher. This suggests that the non-ferrous smelting slag flux provided sufficient

slag volume and oxide dissolution capacity for Nd capture. The Nd concentration remaining in the metal phase was very low in both conditions, with 13 ppm for the fayalite slag flux condition and 11 ppm for the non-ferrous smelting slag flux condition. This indicates that most of the Nd was removed from the metallic phase and transferred into the slag phase regardless of the slag flux type. Therefore, the unrecovered fraction of Nd is unlikely to remain in the metal phase and may instead be associated with slag–crucible interactions, interfacial residues, or mechanical losses during phase separation and sample recovery. Overall, the non-ferrous smelting slag flux showed slightly lower Nd recovery than the synthesized fayalite slag flux, but it still achieved a high recovery rate of 73.16%. These results demonstrate that industrial non-ferrous smelting slag can be used as an alternative flux for pyrometallurgical Nd recovery from end-of-life EV motor materials. The use of such industrial slag by-products may improve the practical applicability and resource circularity of the proposed process by replacing synthetic slag flux with an available secondary material.

3.4. Effect of Crucible Material on Nd Recovery Behavior

Under the optimized conditions determined in the previous sections, Nd was mainly recovered in the slag phase, whereas only trace amounts of Nd remained in the metallic phase. However, the Nd recovery into the slag phase did not reach 100%, suggesting that a portion of Nd may have been retained in other regions, such as the crucible–slag interface or the crucible wall. Therefore, the effect of crucible material on Nd distribution and recovery behavior was investigated. First, the possible interaction between Nd and the alumina crucible was examined thermodynamically. The activity–temperature relationship of the Nd–Al₂O₃ system was calculated using FactSage, as shown in Figure 4. The calculation suggests that Nd can interact with Al₂O₃ and form Nd–Al–O-based complex oxide phases, such as AlNdO₃. This indicates that part of the oxidized Nd may be immobilized at the crucible surface instead of being completely transferred into the slag phase. Therefore, the Nd recovery behavior in this process cannot be explained solely by metal–slag distribution; the interaction between Nd-containing oxide species and the crucible material should also be considered.

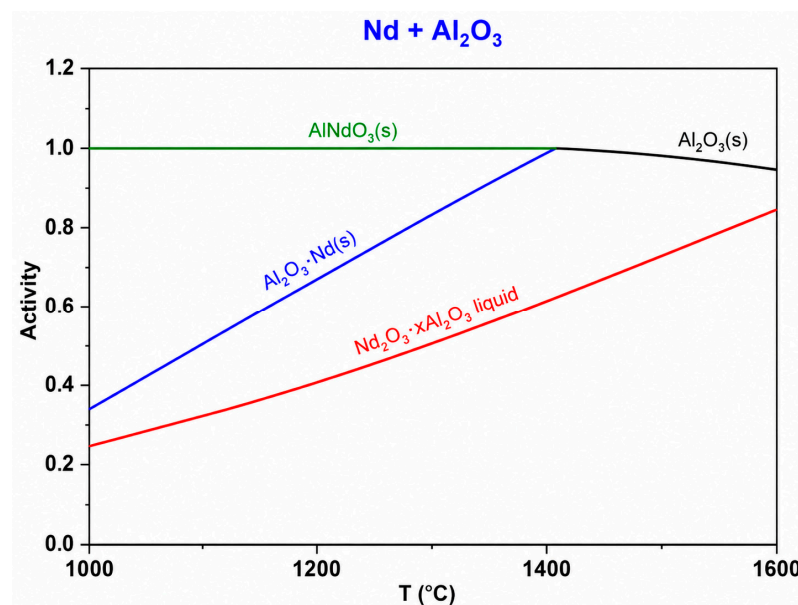


Figure 4. Activity–temperature diagram of the Nd–Al₂O₃ system.

After smelting under the optimized conditions using non-ferrous smelting slag flux, a purple-colored local region was observed on the inner wall of the alumina crucible, as

shown in Figure 5. Since Nd_2O_3 generally exhibits a purple or violet color, this region was considered to be associated with Nd-containing oxide species. The presence of this region at the crucible wall suggests that a portion of Nd was not completely incorporated into the slag phase but remained at the crucible–slag interface or reacted with the crucible surface.

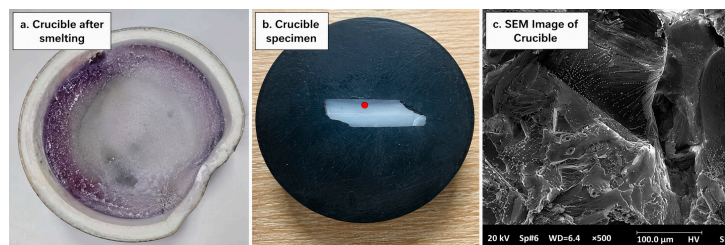


Figure 5. Figure 5. Nd reaction with crucible surface after smelting: (a) crucible after smelting, (b) sectioned specimen, and (c) SEM image of reaction layer. The red circle in (b) indicates the position of the crucible–slag reaction layer selected for SEM–EDS analysis.

To confirm the presence of Nd in this reaction layer, scanning electron microscopy–energy-dispersive X-ray spectroscopy (SEM–EDS) elemental mapping was performed. As shown in Figure 6, Nd was detected in the local reaction layer together with Al and O. This result supports the formation or accumulation of Nd-containing oxide species at the crucible interface. Therefore, the purple-colored region observed after smelting can be attributed to a Nd-enriched reaction layer formed by interaction between Nd oxide and the alumina crucible surface.

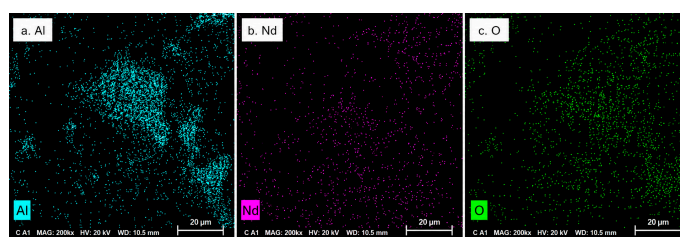


Figure 6. EDS elemental mapping of Nd reaction layer: (a) Al, (b) Nd, and (c) O.

Based on these observations, additional experiments were conducted using different crucible materials to evaluate their influence on Nd recovery behavior. Four types of crucibles were selected: alumina, MgO , zirconia, and carbon crucibles. These crucible materials have different chemical stability, oxide-forming behavior, and interactions with molten slag. The experiments were conducted under identical conditions, using 30 wt% non-ferrous smelting slag flux and 2 wt% Fe_2O_3 under an air atmosphere. The experimental conditions are summarized in Table 11. For clarity, the corresponding material weights based on 100 g of rotor-derived charge are also provided in Table 11.

Table 11. Experimental conditions with different crucible types.

Material/Condition	Value	Weight Based on 100 g Rotor-Derived Charge
Steel	94.32 wt%	94.32 g
Nd–Fe–B magnet	5.68 wt%	5.68 g
Estimated Nd in magnet fraction	1.70 wt%	1.70 g
Total rotor-derived charge	100 wt%	100.00 g
Fe_2O_3 addition	2 wt% of total charge	2.00 g
Non-ferrous smelting slag flux addition	30 wt% of total charge	30.00 g

Atmosphere	Air	–
Crucible type	Alumina, MgO, zirconia, or carbon crucible	–

Note: The listed weights are calculated based on 100 g of rotor-derived charge. The Fe₂O₃ and slag flux additions were calculated relative to the mass of the rotor-derived charge.

The chemical compositions of the recovered slags obtained using different crucible types are summarized in Table 12. The slag composition changed significantly depending on the crucible material, indicating that crucible–slag interaction occurred during high-temperature smelting. These results also show that crucible-derived oxides were partially dissolved or incorporated into the liquid slag phase rather than being completely excluded from the slag.

Table 12. Chemical composition of slag with different crucible types.

Component	Alumina	MgO	Zirconia	Carbon
SiO ₂	50.50	45.25	43.95	48.22
Total Fe as Fe ₂ O ₃ equivalent	11.99	10.70	17.94	18.69
Al ₂ O ₃	25.03	11.80	8.89	12.49
Nd ₂ O ₃	5.92	7.81	6.04	11.91
MnO	0.97	1.10	0.97	0.69
K ₂ O	1.00	1.05	1.16	1.63
ZrO ₂	–	–	13.96	–
Na ₂ O	0.54	–	0.66	–
Cr ₂ O ₃	0.26	–	0.31	0.14
MgO	1.27	19.03	1.76	2.01
CaO	1.85	1.92	2.52	2.62
TiO ₂	0.51	0.45	0.41	0.84
V ₂ O ₅	–	–	0.13	0.10
Dy ₂ O ₃	–	0.37	0.41	0.35
ZnO	0.16	0.06	0.34	0.31
Y ₂ O ₃	–	0.46	0.55	–
Total	100	100	100	100

Note: The Fe content is reported as total Fe expressed as Fe₂O₃ equivalent by XRF analysis. Therefore, the values do not represent the actual FeO/Fe₂O₃ ratio in the recovered slag.

In the alumina crucible, the Al₂O₃ content in the slag was 25.03 wt%, indicating significant incorporation of alumina into the slag. In the MgO crucible, the MgO content increased to 19.03 wt%, confirming the dissolution or incorporation of MgO from the crucible into the slag. Similarly, the zirconia crucible resulted in a ZrO₂ content of 13.96 wt% in the slag, indicating strong interaction between the slag and the zirconia crucible. Therefore, the oxide components derived from the crucible materials were not completely insoluble in the slag. Instead, they were incorporated into the slag to different extents depending on the crucible material. In contrast, the carbon crucible did not introduce a specific refractory oxide component into the slag, and the Nd₂O₃ content in the slag was the highest at 11.91 wt%.

Although crucible-derived oxides were incorporated into the slag phase, the dissolution and transfer of Nd-containing oxide species into the slag were not complete. Part of the Nd was retained in the crucible-reacted region, particularly when oxide crucibles were used. This indicates that Nd-containing oxide species were distributed competitively between the liquid slag phase and the crucible-reacted region. The Nd

distribution among the slag, metal, and crucible-reacted regions was quantitatively evaluated by ICP analysis, and the results are summarized in Table 13. The Nd recovery into the slag phase varied significantly depending on the crucible material. The results indicate that Nd₂O₃ was substantially dissolved or captured in the liquid slag phase, but that the extent of transfer depended on the interaction between Nd-containing oxide species and the crucible material.

Table 13. Effect of crucible type on Nd distribution and recovery.

Crucible Type	Alumina	MgO	Zirconia	Carbon
Total Nd				
Amount of Nd in simulated material (g)	4.21	3	4.08	3.57
Slag phase				
Slag after smelting (g)	86.33	44.15	72.31	60.31
Nd in slag by ICP (ppm)	35,691	49,213	17,494	58,412
Nd in slag (g)	3.08	2.17	1.26	3.52
Nd recovery into slag (%)	73.16 ± 2.19	72.33 ± 2.17	30.88 ± 0.93	98.60 ± 2.96
Metal phase				
Metal after smelting (g)	256.01	189.16	251.43	223.35
Nd in metal by ICP (ppm)	12	89	102	98
Nd in metal (g)	0.003	0.017	0.026	0.022
Nd recovery into metal (%)	0.07 ± 0.01	0.56 ± 0.02	0.63 ± 0.02	0.61 ± 0.02
Crucible-reacted region				
Crucible-reacted region (g)	76.42	71.12	60.11	–
Nd in crucible-reacted region by ICP (ppm)	10,279	7613	46,213	–
Nd in crucible-reacted region (g)	0.79	0.54	2.78	–
Nd loss to crucible-reacted region (%)	18.76 ± 0.56	18.00 ± 0.54	68.14 ± 2.04	–
Overall Nd mass balance				
Overall Nd loss during smelting (g)	1.13	0.81	2.79	0.03
Overall Nd loss during smelting (%)	26.77 ± 2.19	27.11 ± 2.17	68.49 ± 0.93	0.79 ± 2.96
Unaccounted Nd loss after considering crucible-reacted region (g)	0.34	0.27	0.01	0.03
Unaccounted Nd loss after considering crucible-reacted region (%)	8.01	9.11	0.35	0.79

Note: The overall Nd loss was calculated as the difference between the total Nd input and the Nd recovered in the slag and metal phases. The loss term includes unmeasured Nd associated with crucible interaction, adhered slag or metal, incomplete phase separation, sampling loss, and other process-related losses. The uncertainty values were estimated by error propagation using phase mass measurements and a relative ICP uncertainty of 3%.

The complete Nd mass balance shows that the Nd recovered in the metal phase was negligible under all experimental conditions, generally below 1%. Therefore, the difference between the total Nd input and the slag-phase Nd recovery was mainly associated with process-related losses, including crucible interaction, adhered slag or metal, incomplete phase separation, and sampling loss. In the crucible material experiments, the crucible-reacted region was analyzed separately, confirming that crucible interaction was one of the major sources of Nd loss, especially for the zirconia crucible.

The probable mechanism of Nd loss differed depending on the crucible material. In the alumina crucible, Nd loss was mainly attributed to the formation of a Nd-enriched reaction layer at the crucible–slag interface. The SEM–EDS mapping results confirmed that Nd was present together with Al and O in the reaction layer, suggesting the formation or accumulation of Nd–Al–O-based oxide species. This interpretation is consistent with previous phase-equilibrium studies of Nd₂O₃–Al₂O₃- and CaO–Al₂O₃–Nd₂O₃-based

systems, in which Nd-bearing aluminate phases such as NdAlO_3 , CaNdAlO_4 , and $\text{CaNdAl}_3\text{O}_7$ have been reported [13]. Therefore, part of the oxidized Nd may have been immobilized as Nd-containing aluminate species at the alumina crucible interface rather than being completely transferred into the liquid slag phase. In the MgO crucible, the MgO content in the recovered slag increased markedly, indicating partial dissolution or incorporation of MgO from the crucible into the slag. However, the Nd recovery behavior was similar to that of the alumina crucible, and the Nd loss to the crucible-reacted region was slightly lower. This suggests that Nd retention in the MgO crucible was mainly caused by interfacial adhesion and physical or chemical incorporation of Nd-containing oxide species into the crucible-reacted region, rather than by strong immobilization in a specific Nd–Mg–O compound. In contrast, the zirconia crucible showed the most severe Nd loss to the crucible-reacted region. This behavior is attributed to the strong interaction between Nd-containing oxide species and ZrO_2 at the crucible interface. Previous phase-relation and thermodynamic studies of ZrO_2 – Nd_2O_3 -containing systems have shown that rare-earth oxides can interact with zirconia and form Nd–Zr–O or rare-earth zirconate-type phases [22]. In addition, slag–zirconia refractory interaction at high temperature can promote refractory degradation and interfacial reaction layer formation [23]. Therefore, Nd-containing oxide species were strongly retained at the zirconia crucible interface, which explains the low slag-phase Nd recovery and the high Nd loss to the crucible-reacted region. For the carbon crucible, no refractory oxide component was introduced into the slag, and the Nd recovery into the slag phase was the highest. This indicates that Nd was not significantly immobilized by rare-earth oxide–refractory oxide reactions. However, carbon participated in oxidation and reduction reactions in the slag–metal system, causing gas generation, slag foaming, and crucible degradation. Thus, the carbon crucible reduced oxide-type Nd retention but caused severe process instability.

The carbon crucible showed the highest Nd recovery into the slag phase, with 3.52 g of Nd recovered and a recovery rate of 98.60%. This result suggests that Nd was not significantly immobilized by reaction with the carbon crucible and was effectively transferred into the slag phase. However, despite the high Nd recovery, severe process instability was observed during the experiment. As shown in Figure 7, the carbon crucible reacted with the Fe-based molten metal, and slag foaming occurred during smelting. This foaming is likely associated with gas generation caused by carbon oxidation and reduction reactions in the slag–metal system. Therefore, although the carbon crucible exhibited the highest Nd recovery, it is not considered suitable for stable operation because of crucible degradation, carburization of the metal phase, and slag foaming.

The alumina and MgO crucibles showed similar Nd recovery behavior. The Nd recovery into the slag phase was 73.16% for the alumina crucible and 72.33% for the MgO crucible. In both cases, approximately 18% of Nd was detected in the crucible-reacted region, indicating that part of the Nd was immobilized by reaction or interfacial adhesion with the crucible material. Nevertheless, most of the Nd was recovered in the slag phase under these conditions, confirming that Nd-containing oxide species were largely dissolved or captured by the liquid slag rather than remaining entirely at the crucible interface. Although the MgO crucible showed slightly lower crucible-region Nd loss than the alumina crucible, the overall recovery behavior was comparable. Considering cost, availability, and practical handling, the alumina crucible is more advantageous for laboratory-scale and practical process evaluation.

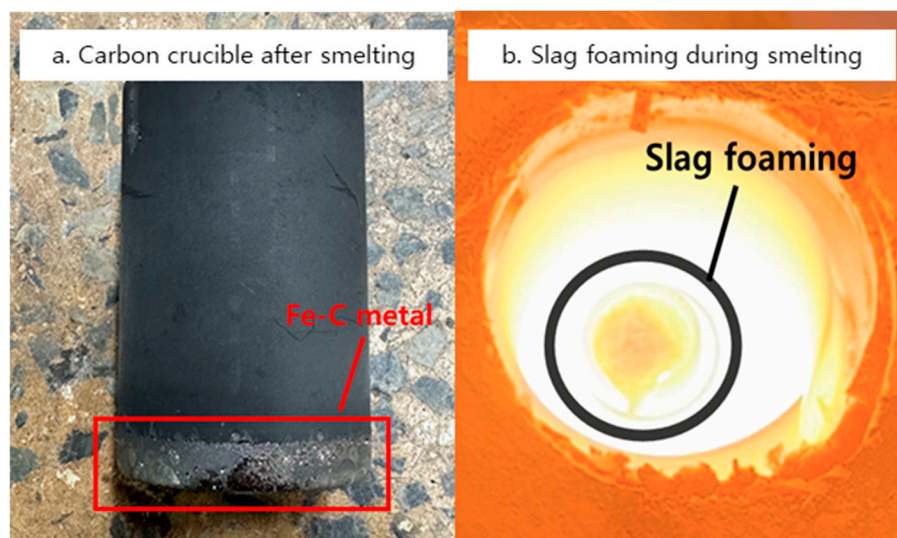


Figure 7. Problems during smelting with carbon crucible: (a) carbon crucible after smelting and (b) slag foaming during smelting.

In contrast, the zirconia crucible showed poor Nd recovery behavior. Only 30.88% of Nd was recovered in the slag phase, whereas 68.14% of Nd was detected in the crucible-reacted region. This indicates that zirconia strongly interacted with Nd-containing oxide species and retained a significant amount of Nd at the crucible interface. Based on the mechanistic interpretation described above, the large Nd loss in the zirconia crucible is most likely associated with the formation of Nd–Zr–O or zirconate-type interfacial reaction products, as well as strong slag–zirconia refractory interaction during high-temperature smelting [22,23]. Therefore, zirconia crucibles are not suitable for the present Nd recovery process.

Although the carbon crucible showed the highest Nd recovery into the slag phase, this result should not be interpreted as indicating that the carbon crucible is the most suitable crucible material for the proposed process. The main objective of this study was Nd recovery into the slag phase; however, the process also produces an Fe-rich metallic phase as a co-product. Therefore, excessive carbon pickup by the Fe-rich metal phase is undesirable because it can deteriorate the quality and recyclability of the metallic product and may require additional downstream decarburization or refining treatment. In addition, carbon dissolution and carbon oxidation/reduction reactions can alter the local oxygen potential of the slag–metal system, which may influence the selective oxidation behavior of Nd and the stability of Nd-containing oxide species in the slag phase. The severe slag foaming observed in the carbon crucible experiment also indicates gas generation during smelting, which can disturb slag–metal separation, increase mechanical entrainment of slag or metal, and reduce process reproducibility. Thus, despite its high apparent Nd recovery, the carbon crucible was considered unsuitable for stable and practical operation because of crucible degradation, carburization of the Fe-rich metal phase, slag foaming, and potential difficulties in downstream treatment.

Overall, the crucible material strongly affected Nd distribution and process stability. Although the carbon crucible provided the highest Nd recovery, severe slag foaming and crucible degradation made it unsuitable for stable operation. The zirconia crucible caused significant Nd loss to the crucible-reacted region. The MgO crucible showed recovery behavior similar to that of the alumina crucible, but the alumina crucible was considered more practical because of its lower cost, availability, and stable handling characteristics. These results indicate that Nd loss was governed by competitive distribution between the liquid slag phase and the crucible-reacted region, depending on the chemical interaction

between Nd-containing oxide species and the crucible material. Therefore, considering Nd recovery, process stability, crucible reactivity, and economic feasibility, the alumina crucible was selected as the most practical crucible material for the present process.

4. Conclusions

In this study, a pyrometallurgical process using non-ferrous smelting slag flux was proposed for the recovery of neodymium (Nd) from Nd–Fe–B magnets contained in end-of-life electric vehicle motors. The process was designed to selectively oxidize Nd in the molten metal phase using Fe_2O_3 and subsequently transfer the oxidized Nd into the slag phase through metal–slag separation. The effects of fayalite slag flux addition, Fe_2O_3 addition, slag flux type, and crucible material on Nd recovery behavior were systematically investigated.

The fayalite slag flux addition ratio strongly affected Nd partitioning into the slag phase. As the slag flux addition increased from 10 to 30 wt%, the Nd recovery into the slag phase increased from 40.12% to 80.92%. This result indicates that sufficient slag volume and oxide dissolution capacity are essential for capturing Nd-containing oxide species. The Nd concentration remaining in the metal phase decreased to a trace level at higher slag flux addition ratios, confirming that Nd was effectively removed from the Fe-based metal phase.

Fe_2O_3 addition was found to be a critical factor for promoting the selective oxidation of Nd. Without Fe_2O_3 addition, the Nd recovery into the slag phase was limited to 36.89%. When the Fe_2O_3 addition was increased to 2 wt%, the Nd recovery sharply increased to 80.92%. Further increases in Fe_2O_3 addition to 3 and 4 wt% did not lead to a meaningful improvement in Nd recovery, indicating that 2 wt% Fe_2O_3 was sufficient to promote Nd oxidation under the present experimental conditions. Therefore, the optimum conditions using the synthesized fayalite slag flux were identified as 30 wt% slag flux and 2 wt% Fe_2O_3 .

The applicability of non-ferrous smelting slag flux as an alternative industrial slag-forming material was then evaluated under the optimized conditions. The non-ferrous smelting slag flux showed a Fe–Si–O-based composition similar to that of the synthesized fayalite slag flux, while also containing additional oxides derived from the industrial smelting process. When non-ferrous smelting slag flux was used, the Nd recovery into the slag phase reached 73.16%. Although this value was slightly lower than that obtained using the synthesized fayalite slag flux, the result demonstrates that industrial non-ferrous smelting slag can be used as a practical alternative flux for Nd recovery.

The crucible material significantly influenced Nd distribution and process stability. The carbon crucible showed the highest Nd recovery of 98.60%; however, severe slag foaming, carburization of the Fe-based metal phase, and crucible degradation were observed during smelting. Therefore, despite its high recovery efficiency, the carbon crucible was not suitable for stable process operation. The zirconia crucible caused significant Nd loss to the crucible-reacted region, resulting in a low slag-phase Nd recovery of 30.88%. In contrast, alumina and MgO crucibles showed similar Nd recovery behavior, with recovery rates of 73.16% and 72.33%, respectively. Considering process stability, cost, availability, and practical handling, the alumina crucible was selected as the most suitable crucible material for the present process.

Overall, the proposed process demonstrated that Nd contained in end-of-life electric vehicle motors can be transferred from motor-derived Fe-based materials into an Nd-bearing slag phase through selective oxidation and metal–slag separation. The use of non-ferrous smelting slag flux provides a practical route for replacing synthetic slag flux with an industrial by-product, thereby improving resource circularity and reducing process cost. However, the Nd recovery obtained in this study is not yet sufficient for direct

industrial implementation as a final process. Therefore, the present work should be regarded as a feasibility study and process-development step for Nd recovery from end-of-life electric vehicle motors. Further studies should focus on improving the overall Nd recovery efficiency through optimization of slag composition, reaction conditions, and crucible materials, as well as the subsequent selective recovery of Nd from the Nd-bearing slag. In particular, selective leaching and conversion of the Nd-bearing slag into high-purity rare-earth compounds such as Nd_2O_3 or NdCl_3 should be further investigated for practical process development and scale-up.

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